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The 8th Trondheim Conference on Capture, Transport and Storage

## Experimental determination of mass-transfer coefficients and area of dumped packing using alkanolamine solvents

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### Abstract

The absorption process is strongly influenced by the effective contact area. In absorber columns, this is related to the type of the internals used in the columns. Therefore, a good representation of the effective mass-transfer area and mass-transfer coefficients ( $k_L$  or  $k_g$ ) is also essential for accurately represent and design a process. For CO<sub>2</sub> capture process packed columns are usually preferred. The mass transfer area and coefficients for several packing (both structured and random) are correlated elsewhere. In this work mass transfer experiments using concentrated MEA solution in the Procede acid gas treating pilot plant are performed. However, due to the fast reaction between MEA and CO<sub>2</sub>, both the gas side and liquid side mass transfer resistances are relevant and the mass transfer area cannot be determined experimentally. Nevertheless, the volumetric mass transfer coefficient is calculated and it showed to be relatively constant for all the runs. The results from the pilot plant are compared to simulated results using Procede Process Simulator (PPS). The mass transfer area and mass transfer coefficients were calculated through the default correlations implemented in PPS. Very good agreement is achieved between the experimental and simulated results.

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## 1. Introduction

CO<sub>2</sub> capture processes have been widely studied and the technology knowledge and employment had increased considerably. Several technologies can be applied for mitigating the CO<sub>2</sub> emissions from flue gases. However, the state of the art is still the absorption with chemical reaction [1, 2]. The first full scale plant for CO<sub>2</sub> post combustion capture, the Saskpower Boundary Dam 1 million tonnes CO<sub>2</sub>/year plant [3], had been recently put into operation and is based on the traditional absorption/desorption process. Despite this notable achievement, there are still more to be done both in terms of research and, mainly, in terms of application.

Several equilibrium and physical properties data had been measured for amine based solvents, especially for the most common solvents such as monoethanolamine (MEA), methyldiethanolamine (MDEA) and piperazine (PZ). The kinetics of CO<sub>2</sub> with many aqueous solutions of alkanolamines is well known. Versteeg et al. (1996) [4] compiled and correlated several experimental data on CO<sub>2</sub> rate of absorption in different aqueous alkanolamines solutions. That information is by all means required for a good interpretation and representation of the process, specifically for process simulation and optimization. Nevertheless, the absorption process is strongly influenced by the effective contact area. In absorber columns, this is related to the type of the internals used in the columns. Therefore, a good representation of the effective mass-transfer area and mass-transfer coefficients ( $k_L$  or  $k_g$ ) is also essential for accurately represent and design a process.

### Nomenclature

$a_e$	Effective mass transfer area, m <sup>2</sup> /m <sup>3</sup>
DEA	Diethanolamine
$E$	Enhancement factor
$E_{CO_2}^\infty$	Infinite enhancement factor
$Ha$	Hatta Number
$H_{CO_2}$	Henry's constant, kPa.m <sup>3</sup> .mol <sup>-1</sup>
$K_G$	Overall mass transfer coefficient, mol/(s.m <sup>2</sup> .kPa) or m/s
MEA	Monoethanolamine
PPS	Procede Process Simulator
$R$	Universal gas constant, kPa.m <sup>3</sup> /(mol.K)
$T$	Temperature, K
$u_G$	Superficial gas velocity, m/s
$Z$	Packing height, m

For CO<sub>2</sub> capture process packed columns are usually preferred. The mass transfer area and coefficients for several packing (both structured and random) are correlated elsewhere [5, 6, 7].

To validate a model, pilot plant data is often employed. Reported tests on pilot plants usually shows the energy requirements and/or the best operational conditions for a particular solvent [8, 9, 10]. Those types of data can as well be used for model validation. When the simulation does not match the experimental results, the mass transfer area and/or the mass transfer parameters are generally optimized. This is usually done because the correlations used to calculate mass transfer area and parameters uses data generated in different conditions as the absorption process is applied. For instance, the solvent is usually diluted. Contrary to other studies [11, 12, 13] this work performed mass transfer experiments using concentrated MEA solution in the Procede acid gas treating pilot plant. However, due to the fast reaction between MEA and CO<sub>2</sub>, both the gas side and liquid side mass transfer resistances are relevant and the mass transfer area cannot be determined experimentally. Nonetheless, the volumetric mass transfer coefficient is calculated and it showed to be relatively constant for all the runs.

The results from the pilot plant are compared to simulated results using Procede Process Simulator (PPS). The mass transfer area and mass transfer coefficients were calculated through the default correlations implemented in PPS. Very good agreement is achieved between the experimental and simulated results.

## 2. Theoretical background

A common way to model the performance of an absorber is through a series of resistances as expressed in Eq. 1.

$$\frac{1}{K_G} = \frac{1}{k_g} + \frac{H_{CO_2}}{Ek_l^\circ} = \frac{1}{k_g} + \frac{1}{k_g'} \quad 1$$

If the experiment is carried out in the so-called pseudo-first order regime, the following condition must hold [14]:

$$2 < Ha \ll E_{CO_2}^\infty \quad 2$$

Eq. 3 expresses the Hatta number for the pseudo-first order reactions involving MEA and CO<sub>2</sub>. The infinite enhancement factor expression will depend on the theoretical model and it's given elsewhere for the different models. In this work, the reaction rate constant and the CO<sub>2</sub> diffusion coefficient are calculated according to [4].

$$Ha = \frac{\sqrt{k_{MEA}[MEA]D_{CO_2}}}{k_l^\circ} \quad 3$$

When the pseudo-first order regime is attained, the enhancement factor can be considered equal to the Hatta number, and

$$k_g' = \frac{\sqrt{k_{MEA}[MEA]D_{CO_2}}}{H_{CO_2}} \quad 4$$

The expression for the volumetric mass-transfer coefficient (Eq. 5) is derived from the differential mass balance along the absorber [12].

$$K_G a_e = \frac{u_G \ln \left( \frac{y_{CO_2, in}}{y_{CO_2, out}} \right)}{ZRT} \quad 5$$

When the experiment is carried under certain conditions, the gas-side resistance may be neglected and, as seen from Eq. 1, the overall mass transfer coefficient equals the parameter  $k_g'$ . Once the overall mass transfer coefficient is known, the effective interfacial area of the packing can be calculated through Eq. 5.

## 3. Procede pilot plant

Figure 1 shows the simplified flow diagram from the Procede pilot plant. The plant design is based on the traditional absorption/desorption process [15]. From a 3 kW blower air is blown to a humidifier to saturate the gas. Prior to the humidifier inlet, pure CO<sub>2</sub> is added to the stream until a desired CO<sub>2</sub> concentration is achieved in the gas stream. From the humidifier, the saturated gas enters the bottom of the absorber where it meets the lean solvent which flows countercurrent. The rich solvent leaving at the bottom of the absorber is sent to the regeneration section while the low CO<sub>2</sub> concentration gas leaving at the top of the absorber is vented to the atmosphere. The gas flow can be measured between 40-140 m<sup>3</sup>/h via the calibrated vortex gas flow meter. This corresponds to a superficial gas velocity of approximately 0.3-1.2 m/s.

The rich solvent is sent to the top of the desorber after passing through a cross flow heat exchanger. The energy required for regenerating the solvent is provided by an electrical reboiler. The lean solvent is sent back to the absorber passing through the cross flow heat exchanger and a cooler before entering at the top of the absorber. The vapour leaving the top of the desorber is passed through a cooler where water and solvent are condensed. The condensate is returned to the system while the resulting gas (almost pure CO<sub>2</sub>) is released to the atmosphere.

The absorber consists of a transparent 8.5 meters high casted Poly(methyl methacrylate) (PMMA) column with an internal diameter of 175 mm and a wall thickness of 5 mm. The absorber is filled with 16 mm polypropylene (PP) Pall rings up to a height of 6.85 m. At 1.31, 2.76, 4.21, 5.66, and 6.85 m from the top, sample points for liquid and

gas are available. Additionally, temperature indicators are installed and the liquid flow is redistributed towards the center of the packing.

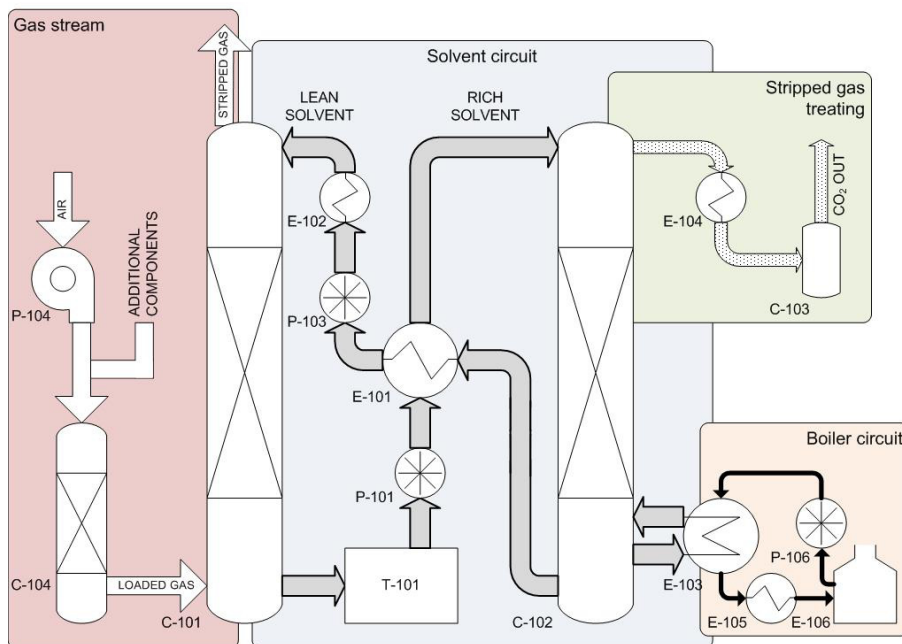


Figure 1: Procede pilot plant simplified flowchart [16].

The desorber is a double walled 8.5 meters high 304 stainless steel column with 100 mm internal diameter and 3 mm wall thickness. A Mellapak 250Y structured packing is installed with a bed height of 6.92 m. Similar to the absorber, five temperature indicators are installed at different packing depths, together with one temperature indicator on the desorber sump. A pressure indicator is installed at a packing depth of 5.03 m.

#### 4. Experimental procedure

Approximately 180 liters of a 30 mass % MEA aqueous solution was gravimetrically prepared and filled into the pilot. A liquid and gas flow was set and CO<sub>2</sub> was added to the blown air controlled by a pneumatic valve so that its inlet concentration was about 2% vol. The inlet and outlet gas concentration were continuously sampled. Liquid samples were taken at the lean loading line before the lean solvent enters the absorber and right after the absorber sump (rich line). Once the temperatures, pressures and flows were stable, the pilot was left for at least 2 hours running before liquid samples were taken. After the samples were taken, a new condition was set by mainly varying the liquid and/or gas flow. The CO<sub>2</sub> concentration in the gas was measured at absorber inlet and outlet conditions through an IR CO<sub>2</sub> analyzer. The gas flow was varied from 55 to 85 m<sup>3</sup>/h while the liquid flow was varied from 180 to 400 kg/h. The reboiler temperature was set to 125 °C for all runs. A total of 19 experimental points were generated.

The CO<sub>2</sub> absorbed from the gas was calculated in two different ways: mass balance based on the gas and liquid streams. Acid-base titration was used to determine the amine concentration in the liquid samples. The CO<sub>2</sub> concentration in the liquid samples was determined via non-aqueous titration [17, 18] in a Procede customized apparatus.

## 5. Results and discussion

The infinite enhancement factor was calculated using the film theory [4]. The Hatta number was calculated using the kinetic parameters from [4], the liquid side mass transfer coefficient model from [5] and physical properties calculated from default PPS correlations [19]. The experiments were carried under the pseudo-first order regime given that all experiments fulfill Eq. 2.

The deviation between the mass balance calculated from the gas and liquid streams are within 10%. For the comparison with PPS, the experimental absorbed  $\text{CO}_2$  is calculated from the gas side.

PPS was used to simulate the experimental runs and the gas inlet and lean solvent experimental conditions were used as inputs in PPS to simulate the pilot plant absorber. The experimental and simulated outputs of the absorber (rich solvent and clean gas) were compared. PPS uses a rigorous rate based model to calculate the absorber performance. All mass transfer and kinetic correlations used were the default correlations in PPS. The interfacial area and the liquid side mass transfer coefficient were calculated using Billet and Schultes correlation [5] while the gas side mass transfer coefficient was calculated using Onda and coworkers correlation [6]. Figure 2 shows the good agreement between the simulated and experimental absorbed  $\text{CO}_2$ . It's also seen that PPS mainly under predicts the amount of  $\text{CO}_2$  capture. Nevertheless, this under prediction is within 10%.

The agreement between the experimental and simulated rich loadings was excellent. Except for two runs the agreement was within 5% as shown in Figure 3. The liquid flow rate for the rich stream presented an even better agreement. Except for two runs, all runs showed deviations between experimental and simulated below 1.5 %. From these observations, it's possible to conclude that PPS is able to satisfactorily represent the pilot plant for the MEA campaign using its default correlations. Figure 2, nevertheless, may indicate that the interfacial area should be slightly increased to allow more capture.

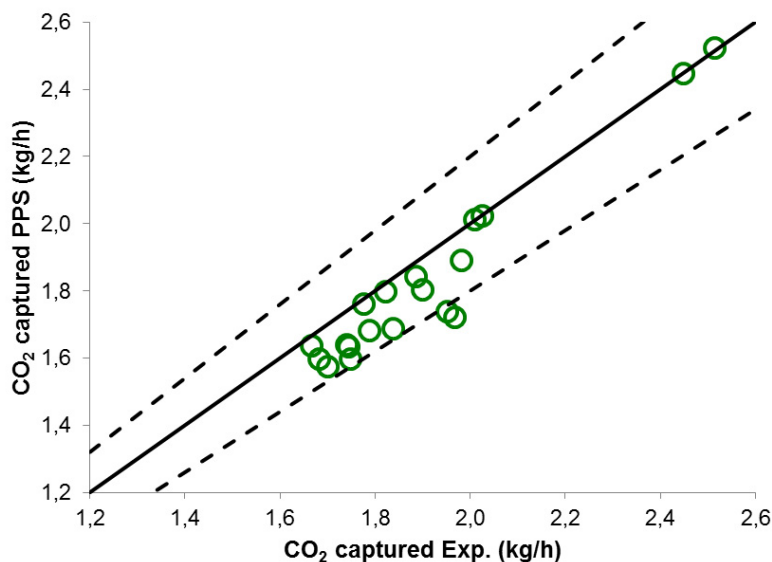


Figure 2: Parity plot of simulated and experimental  $\text{CO}_2$  captured. (O) Experimental points, (—)  $x = y$ , (---)  $\pm 10\%$ .

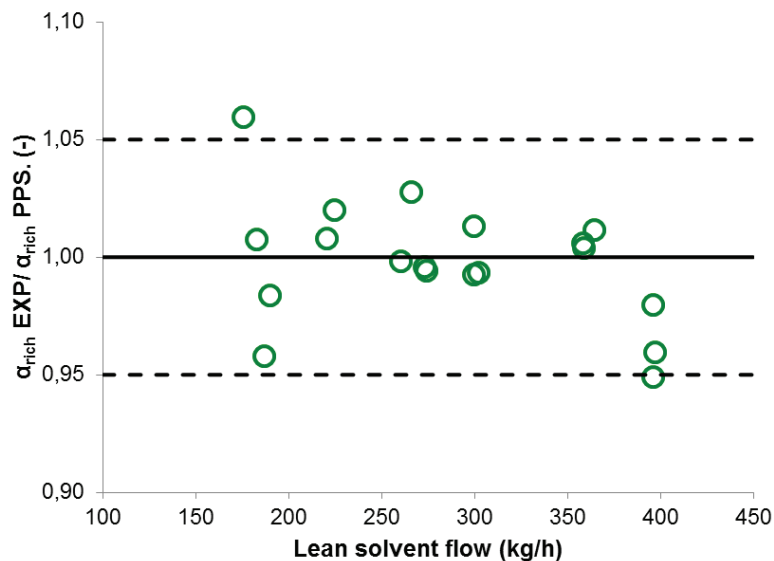


Figure 3: Rich loading deviation.

In PPS simulations no heat loss was assumed. In the acid gas treating pilot plant the absorber is made from a transparent material and it's not thermally isolated. So, it's expected to have a significant heat loss as seen in Figure 4. Nevertheless, it's still possible to see the expected trend in the temperature profile for the absorber where the middle of the column is warmer than the top and bottom.

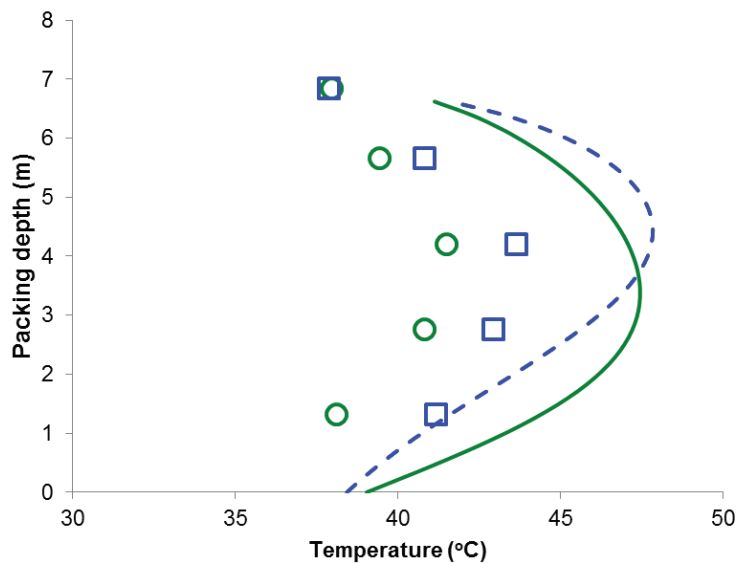


Figure 4: Absorber temperature profile. Experimental: (●) run 14 and (□) run 19, PPS model: (—) run 14 and (---) run 19

For the experiments performed, both gas and liquid side mass-transfer resistances (Eq. 1) are relevant and none can be neglected, to simplify the calculation of the overall mass-transfer coefficient. This is mainly due to the fast kinetics between MEA and CO<sub>2</sub>. With that, the interfacial area cannot be isolated from the overall mass transfer coefficient in Eq. 5.

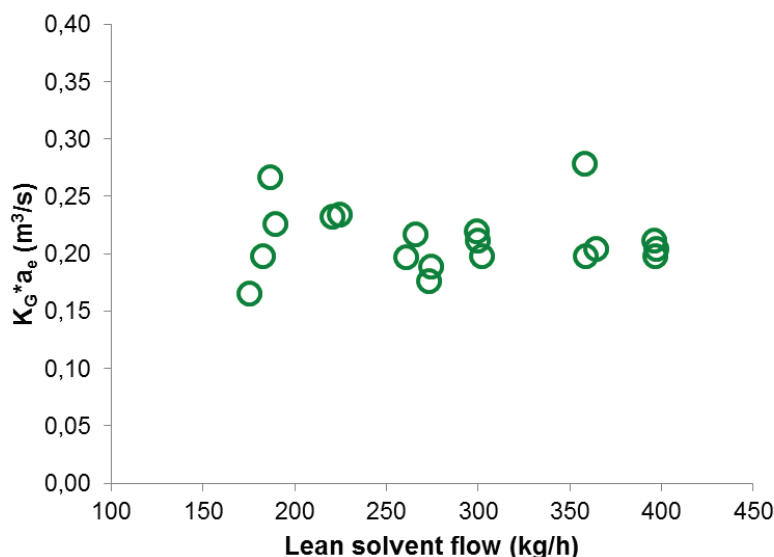


Figure 5: Volumetric mass-transfer coefficient as function of the lean solvent flow.

Figure 5 and 6 show the volumetric mass-transfer coefficient for all runs in the pilot plant calculated with Eq. 5. The volumetric mass-transfer coefficient seems to be constant at around 0.2 m/s for all runs. From the default correlations implemented in PPS, the simulated volumetric mass-transfer coefficients are also calculated. The agreement between the experimental and calculated volumetric mass-transfer coefficients were within 20%, except for a few measurements. The calculated volumetric mass transfer coefficient was higher than the experimental for most of the runs as shown in Figure 7.

The results shown in Figure 7 indicate that there is a strong interaction between the liquid side mass transfer coefficient, gas side mass transfer coefficient and interfacial mass transfer area which keeps the volumetric mass transfer coefficient relatively constant. One interpretation from Figure 2 is that the mass transfer area should be greater than what is calculated by the correlation. By increasing the mass transfer area, the amount of absorbed CO<sub>2</sub> is also increased if all other coefficients are kept constant. In Figure 7, it's seen that PPS estimates a higher volumetric mass transfer parameter. If the area is increased and all other parameters are kept constant, the deviation between the estimated and experimental volumetric mass transfer parameter shall increase, indicating that there is a change in other mass transfer parameters which keeps the volumetric mass transfer parameter constant.

The mass transfer parameters are usually estimated using a general correlation as, for example, in [5, 6, 7]. These correlations are, as mentioned, general and were estimated outside the process conditions for the CO<sub>2</sub> capture process. A common approach in simulations is to fine tune the mass transfer parameters using pilot plant data, so that the calculated results match the experimental data. The experimental results given in this work, therefore, can contribute to validate the mass transfer parameters correlations. Although the comparison cannot be directly with the individual mass transfer parameters, the results from this work can be used to optimize the volumetric mass transfer coefficient and, thus, increasing the understanding of the process and simulation accuracy.



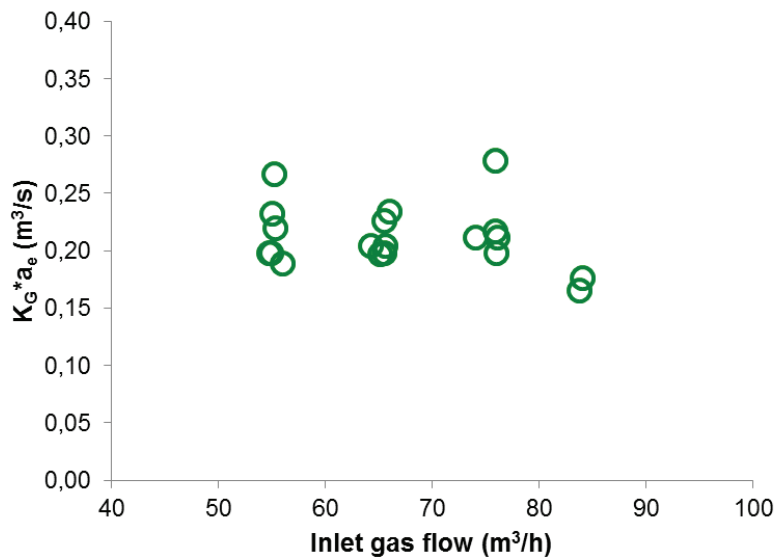


Figure 6: Volumetric mass-transfer coefficient as function of the inlet gas flow

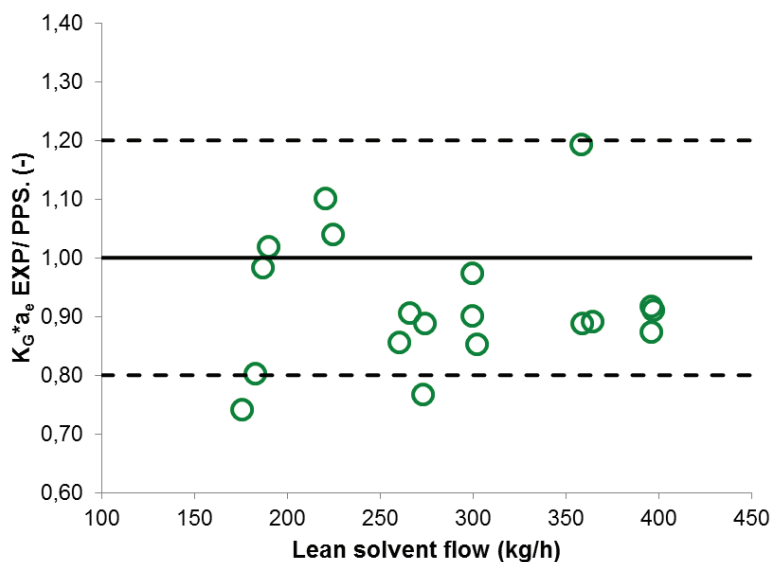


Figure 7: Volumetric mass-transfer parameter comparison.

## 6. Conclusions and recommendations

The Procede pilot plant was used to determine the volumetric mass transfer coefficient of the Pall rings dumped packing used in its absorber. A 30 wt. % MEA aqueous solution was used as the solvent. The mass balance was

calculated in two ways, using the gas and liquid analyses. The two procedures are in good agreement and the mass balance calculated through the gas analysis was chosen to be used for further comparisons.

PPS was used to simulate the pilot plant runs using its default correlation. The inlets of the absorber (gas and lean solvent) were assumed to be known from the experimental results and the output of the absorber were calculated. The agreement between the simulated results and the experimental was very good, especially for the rich solvent flow and rich loading. The amount of CO<sub>2</sub> captured calculated through PPS was usually lower than the experimental suggesting that the calculated mass transfer area should be increased.

Due to the conditions of the experiments, both the liquid side and gas side mass transfer resistances are relevant and none can be neglected to simplify the calculation of the overall mass transfer coefficient. Thus, the mass transfer area cannot be experimentally determined, but instead the volumetric mass transfer coefficient. The experimental volumetric mass transfer coefficient was rather constant for all 19 runs. The calculated volumetric mass transfer coefficient using PPS was in good agreement with the experimental ones.

To independently measure the mass transfer area and coefficients, the experiment must be performed so that one can eliminate the effect of one of the mass transfer resistances. With real process conditions, aqueous solution of MEA is not suited for this type of experiment due to its fast kinetics with CO<sub>2</sub>. Tests with aqueous solution of DEA will be performed to determine independently the mass transfer area of the absorber packing.

## References

- [1] H. F. Svendsen, E. T. Hessen and T. Mejdell, "Carbon dioxide capture by absorption, challenges and possibilities," *Chemical Engineering Journal*, vol. 171, pp. 718 - 724, 2011.
- [2] G. T. Rochelle, "Amine Scrubbing for CO<sub>2</sub> Capture," *Science*, vol. 325, pp. 1652-1654, 2009.
- [3] SaskPower, "Boundary dam integrated carbon capture and storage demonstration project," 2012. [Online]. Available: [http://www.saskpower.com/wp-content/uploads/clean\\_coal\\_information\\_sheet.pdf](http://www.saskpower.com/wp-content/uploads/clean_coal_information_sheet.pdf). [Accessed 21 March 2014].
- [4] G. F. Versteeg, L. A. J. van Dijck and W. P. M. van Swaaij, "On the kinetics between CO<sub>2</sub> and alkanolamines both in aqueous and non-aqueous solutions. An overview," *Chemical Engineering Communications*, vol. 144, pp. 113-158, 1996.
- [5] R. Billet and M. Schultes, "Prediction of Mass Transfer Columns with Dumped and Arranged Packings: Updated Summary of the Calculation Method of Billet and Schultes," *Chemical Engineering Research and Design*, vol. 77, pp. 498 - 504, 1999.
- [6] K. Onda, H. Takeuchi and Y. Okumoto, "Mass transfer coefficients between gas and liquid phases in packed columns," *Journal of Chemical Engineering of Japan*, vol. 1, pp. 56 - 62, 1968.
- [7] J. L. Bravo and J. R. Fair, "Generalized correlation for mass transfer in packed distillation columns," *Industrial & Engineering Chemistry Process Design and Development*, vol. 21, pp. 162 - 170, 1982.
- [8] R. Notz, H. P. Mangalapally and H. Hasse, "Post combustion CO<sub>2</sub> capture by reactive absorption: Pilot plant description and results of systematic studies with MEA," *International Journal of Greenhouse Gas Control*, vol. 6, pp. 84 - 112, 2012.
- [9] F. A. Tobiesen, O. Juliussen and H. F. Svendsen, "Experimental validation of a rigorous desorber model for post-combustion capture," *Chemical Engineering Science*, vol. 63, pp. 2641 - 2656, 2008.
- [10] D. D. D. Pinto, H. Knuutila, G. Fytianos, G. Haugen, T. Mejdell and H. F. Svendsen, "CO<sub>2</sub> post combustion capture with a phase change solvent. Pilot plant campaign," *International Journal of Greenhouse Gas Control*, vol. 31, p. 153-164, 2014.
- [11] C. Wang, M. Perry, G. T. Rochelle and A. F. Seibert, "Packing Characterization: Mass Transfer Properties," *Energy Procedia*, vol. 23, pp. 23 - 32, 2012.
- [12] R. E. Tsai, A. F. Seibert, R. B. Eldridge and G. T. Rochelle, "A dimensionless model for predicting the mass-transfer area of structured packing," *AIChE Journal*, vol. 57, pp. 1173-1184, 2011.
- [13] A. Zakeri, A. Einbu, P. O. Wiig, L. E. Øi and H. F. Svendsen, "Experimental investigation of pressure drop,

liquid hold-up and mass transfer parameters in a 0.5 m diameter absorber column," *Energy Procedia*, vol. 4, pp. 606 - 613 , 2011.

- [14] P. Danckwerts, *Gas-Liquid Reactions*, New York: McGraw-Hill Inc, 1970.
- [15] R. R. Bottoms, " Process for separating acidic gases," 1930.
- [16] S. Bruggink, *Acid Gas Treating Pilot Plant: Auditing, Optimization and Characterization*, University of Groningen, 2013.
- [17] L. Blom and L. Edelhausen, "Direct titration of carbon dioxide," *Analytica Chimica Acta*, vol. 13, pp. 120 - 128, 1955.
- [18] R. F. Jones, P. Gale, P. Hopkins, Powell and L. N., "A Simple and Rapid Titrimetric Method for the Determination of Carbon in Iron and Steel," *Analyst*, vol. 90, pp. 623 - 629, 1965.
- [19] E. P. van Elk, A. R. J. Arendsen and G. F. Versteeg, "A new flowsheeting tool for flue gas treating," *Energy Procedia*, vol. 1, p. 1481–1488, 2009.